

The local magnetic moments and hyperfine magnetic fields in disordered metal-metalloid alloys

A.K.Arzhnikov and L.V.Dobysheva

Physical-Technical Institute, Ural Branch of Russian Academy of Sciences,
Kirov str. 132, Izhevsk 426001, Russia

The local magnetic moments and hyperfine magnetic fields (HFF) in the ordered alloys $Fe_{15}Sn$ and $Fe_{15}Si$ are calculated with the first-principles full-potential linear augmented plane wave (FP LAPW) method. The results are compared with the experimental data on $Fe - M$ ($M = Si, Sn$) disordered alloys at small metalloid concentration. The relaxation of the lattice around the impurity and its influence on the quantities under consideration are studied. The mechanism of the local magnetic moment formation is described. It is proved that the main distinction between these alloys is connected with the different lattice parameters. Three contributions to the HFF are discussed: the contributions of the core and valence electron polarization to the Fermi-contact part, and the contribution from the orbital magnetic moment.

75.50.Bb, 71.15.Ap

This paper deals with the low-concentration dependences of the local magnetic moments and hyperfine magnetic fields (HFF) at the Fe nuclei for the most typical metal-metalloid alloys $Fe_{1-x}Si_x$ and $Fe_{1-x}Sn_x$.

The first-principles calculations were performed for the ordered alloys $Fe_{15}Sn$ and $Fe_{15}Si$ with lattices obtained by substituting a metalloid atom for one of the Fe atoms in the extended BCC cell of 16 atoms. The unit cell contains 4 non-equivalent Fe atoms located at different distances from the metalloid atom (Fig. 1). Such a model of the alloy was chosen in accordance with the experimental data [1,2] which show that the BCC structure of the substitution alloys is retained up to 30at.% of metalloid. Fig. 2 displays the BCC structure parameter taken for $Fe - Si$ from [2,5,6] and for $Fe - Sn$ from [7]. The self-consistent band structure calculations were performed using the full-potential linearized augmented plane waves (FLAPW) method with the WIEN97 program package [3,4].

The experimental lattice constants taken for the impurity concentration of 6.25at.% (Fig. 2) and extrapolated to the zero temperature were used (for $Fe_{15}Sn$ $a_{Sn} = 10.9924a.u.$, for $Fe_{15}Si$ $a_{Si} = 10.7926a.u.$). To illustrate the lattice parameter effect, we carried out the calculations for $Fe_{15}Sn$ with $a_{Fe} = 10.8114a.u.$ corresponding to pure Fe . For $Fe_{15}Sn$ and $Fe_{15}Si$ the relaxation was studied by shifting the iron atoms FeI closest to the metalloid atom along the main diagonal of the cube. The minimum total energy was obtained for a shift $\delta r = 0.008a_{Sn}\sqrt{3}$ for tin and $\delta r = -0.001a_{Si}\sqrt{3}$ for silicon, which corresponds to the experimental tendency of the lattice expansion/contraction in the case of Sn/Si (Fig. 2). This indicates that a slightly distorted BCC structure should occur in reality (see Fig. 1).

Hereafter the "local magnetic moment" means the integrated spin density of the d -like electrons in the muffin-tin sphere M_d . Fig. 3 shows the local magnetic moments at the Fe atoms as a function of the distance to the metalloid atom in the ordered alloys $Fe_{15}M$ ($M = Sn, Si$) for the non-relaxed and relaxed structure. The M_d mag-

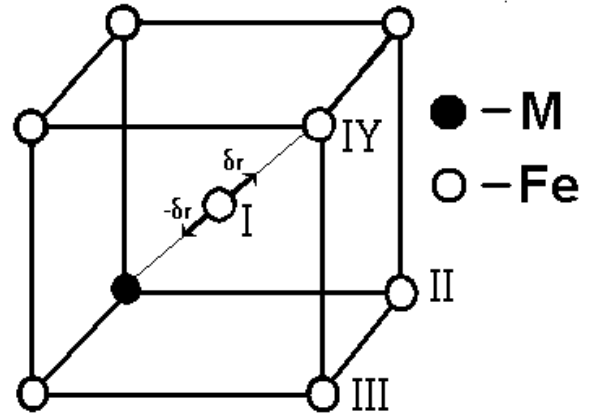


FIG. 1. Unit cell of the ordered alloy $Fe_{15}M$

nitude is governed by two opposite factors. The first is connected with the effective overlap of the d -like wave functions that decreases with the interatomic distance or impurity amount. The second deals with the d -band flattening with the increase of the $s - d$ hybridization at an iron site due to the potential distortion by impurity. The local character of the $s - d$ hybridization results in the fact that this effect takes place only near the impurity. The distinctions in the local magnetic moments at the equivalent positions between the alloys with Si and Sn are a result of different inter-atomic distances. To prove this, the analysis of the $Fe_{15}Sn$ alloy with the lattice parameter of the $Fe_{15}Si$ system was conducted. The M_d values in alloys with Si and Sn were close if the lattice parameters were taken equal. The M_d values increase with the lattice parameter, which is also confirmed by the calculations for pure iron with different lattice parameters. One can see (Fig. 3) that decreasing the distance from FeI to the nearest neighbours in the relaxed system $Fe_{15}Sn$ results in a decrease of the magnetic moment which approaches that of pure iron. We should note that

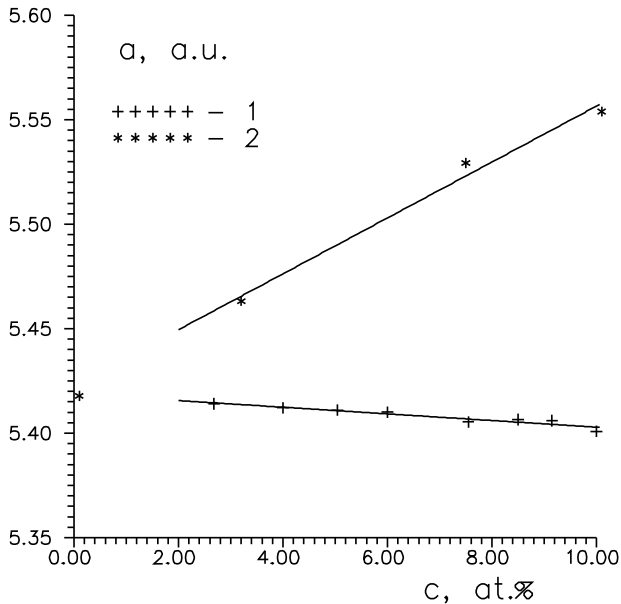


FIG. 2. Concentrational dependence of the BCC lattice parameter for the disordered alloys $Fe-Si$ (1) and $Fe-Sn$ (2)

the distance to the farther spheres determined by the lattice parameter remains completely unchanged or, for some spheres, unchanged on the average, thus, the $FeIY$ magnetic moment is governed mainly by the distance to the nearest neighbours. The difference in the total magnetization for the systems with Si and Sn corresponds to the experimental value at the concentration 6.25at.%.

The electron-polarization contribution to the HFF was calculated by the standard procedure of electron spin-density integration with the relativistic effects taken into account [8]. This contribution includes two terms: the polarization of the inner-levels electrons ("core" electrons) in the nucleus region H_{cor} and that of the valence electrons H_{val} . The polarization of the core electrons follows the relation $H_{cor} = \gamma M_d$ closely. Usually, H_{cor} is considered to be proportional to the total magnetization, which is much worse. The factor γ depends on the type of the exchange-correlation potential. On the whole, the calculations were done with the generalized gradient approximation (GGA) of the potential [9], this gave $\gamma = -123kG/\mu_B$. The calculations with the local-density approximation (LDA) of the potential [10] change γ to $-112kG/\mu_B$. However, within the approximation chosen, γ keeps constant with an accuracy of one percent for different alloys (with Si or Sn), different lattices (BCC, hexagonal), different lattice parameters. The dependence of H_{val} on the distance to the impurity has an oscillatory shape and resembles the Ruderman-Kittel-Kasuya-Yosida (RKKY) polarization. As shown in [11], the simple function $\cos(2K_F r)/r^3$ obtained in the model of free-electron polarization reflects the main features of the RKKY polarization in these alloys, i.e. the period, amplitude, phase; but it does not take into consideration

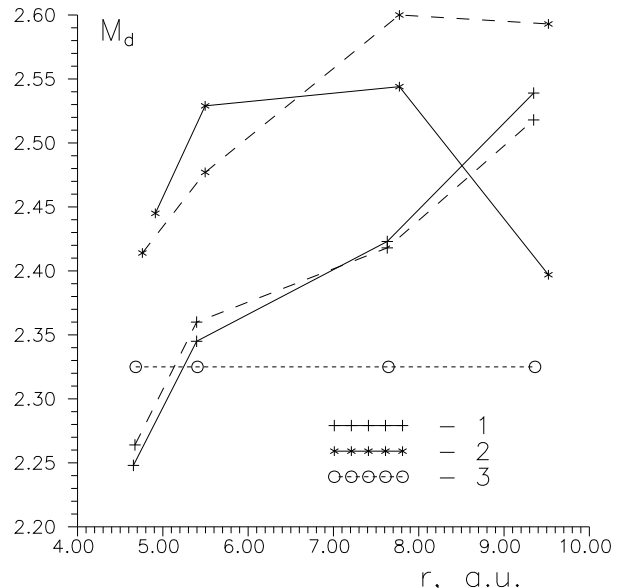


FIG. 3. Local magnetic moments (μ_B) (integrated spin density of the d-like electrons) at Fe atoms in four non-equivalent positions, 1 - $Fe_{15}Si$, 2 - $Fe_{15}Sn$, 3 - pure Fe . Dashed/solid lines are guidelines for values in non-relaxed/relaxed lattice.

the spatial distribution of the $s-d$ exchange interaction and inhomogeneity of the valence-electron density, and so does not allow a quantitative analysis.

A large contribution to the HFF is due to the orbital magnetic moment M_{orb} . In pure iron, M_{orb} resulting from the reduction of the Hamiltonian symmetry by the spin-orbit correction is of $0.08\mu_B$. With the metalloid impurity inclusion, the cubic symmetry of the crystal potential is also violated, which gives an additional defreezing of the orbital magnetic moment, and its growth leads to a decrease in the HFF absolute value. This is corroborated by almost equal experimental differences $H_0 - H_1 \approx 20kG$ for both $Fe_{100-c}Si_c$ [12] and $Fe_{100-c}Sn_c$ [13] (H_0 is the HFF at the nucleus of Fe without metalloid atoms in the nearest environment, and H_1 is that with one metalloid atom in the nearest environment). This difference could not be explained by either change of the core electron polarization (H_{cor}) due to the magnetic moment change (Fig. 3), or the change in the RKKY polarization (H_{val}) as was shown in [11]. We would like to remind that slightly increasing the orbital magnetic moment by $\approx 0.06\mu_B$ should give a decrease of $20kG$ in the HFF absolute value [14]. Though present first-principles calculations do not give exactly the experimental values of the orbital magnetic moment, our computations [15] showed that the orbital magnetic moment and its contribution to the HFF increase when an impurity appears in the nearest environment of an iron atom.

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